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AN APPARATUS FOR APPLYING THE TECHNIQUE OF
ELECTROCHEMILUMINESCENCE TO BOUNDARY
LAYER STUDIES

by

Lt. THOMAS RICHARD SCHILLER, USCG

SUBMITTED TO THE DEPARTMENT OF NAVAL ARCHITECTURE AND MARINE
ENGINEERING IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR
THE MASTER OF SCIENCE DEGREE IN MECHANICAL ENGINEERING
AND THE PROFESSIONAL DEGREE, NAVAL ENGINEER

at the
MASSACHUSETTS INSTITUTE OF
TECHNOLOGY

May, 1964

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Monterey, California

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Signature of Author
Department of Naval Architecture and
Marine Engineering, May 22, 1964

Certified by
Thesis Supervisor

Accepted by
Chairman, Departmental Committee
on Graduate Students

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Thesis

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ABSTRACT

AN APPARATUS FOR APPLYING THE TECHNIQUE OF
ELECTROCHEMILUMINESCENCE TO BOUNDARY LAYER STUDIES

by

Lt. THOMAS RICHARD SCHILLER, USCG

Submitted to the Department of Naval Architecture and Marine Engineering on May 22, 1964 in partial fulfillment of the requirements for the Master of Science Degree in Mechanical Engineering and the Professional Degree, Naval Engineer.

The technique of electrochemiluminescence has been studied as a possible aid in the study of boundary layers. A platinum coated body is placed in a flowing stream of chemiluminescent solution and when a potential is applied, a blue glow appears on the surface of the body. The amount of glow is governed by the rate of arrival of reactants at the surface of the body. Variations in the intensity of glow in different areas suggests the probability that the glow pattern is coupled to the flow in the boundary layer over the surface.

A "blowdown" system was constructed, consisting of a large settling tank approximately twenty feet above the

discharge, a contraction cone, a test section four inches in diameter, and a catch tank. The system parameters allow a maximum velocity in the test section of ten feet per second which corresponds to a Reynolds number of 10^6 measured at one foot of length in the test section.

Preliminary experiments were conducted in a small apparatus to determine how the basic solution can be varied and still obtain satisfactory results for visual studies which are not involved in light intensity measurements. The results of the experiments were used in full scale tests in the large apparatus.

A qualitative study was performed to show the variation of the position of separation on a flat plate with Reynolds number and the angle of attack as variables. Another test was run to show that electrochemiluminescence is useful in the study of turbulence stimulators used in ship model resistance tests.

Thesis Supervisor: George S. Springer

Title: Assistant Professor of Mechanical Engineering

ACKNOWLEDGEMENTS

The author wishes to express his appreciation to Professor George S. Springer, Department of Mechanical Engineering, Massachusetts Institute of Technology for providing the topic of Electrochemiluminescence and for his guidance and many suggestions which resulted in continued interest in the subject without which, the final goal could not have been reached; to the technicians in the Engineering Projects Laboratory for their able assistance in the construction of the apparatus and to Lt. John Ulrich, USN for his time and effort spent helping bring the project to completion.

This project was supported in part by the United States Navy, whose financial aid is gratefully acknowledged.

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I. INTRODUCTION

A. Background

Methods of visualization of flow phenomena are an important part of the study of the science of fluid mechanics. Such techniques have proven useful not only to substantiate hypotheses or to verify existing theories, but to provide a means of discovery of new fluid flow phenomena. It is a known fact that the discovery of certain fluid phenomena, such as the existence of the turbulent regime by Reynolds, resulted from a visual study (1)*.

With the aid of visual studies, we can expect to link a purely "mathematical model" to an actual flow pattern and to possibly identify definite flow fields such as the position of transition from laminar to turbulent flow and the position of separation of flow. With the aid of the visual study, we can define the limits of the governing parameters over the specified regimes.

Flow visualization is also of prime interest and importance in the field of engineering applications. Since the governing equations of fluid mechanics are particularly non-linear and very few exact solutions exist today, we

* Numbers in parentheses correspond to references as listed in the bibliography.

would be seriously hampered in the development of new engineering systems if we could not depend on methods of flow visualization to aid in predicting possible flow patterns.

Through the years, interest in the field of flow visualization has led to the development of several techniques which include:

- a. Smoke Filaments
- b. Dye Injection
- c. Neutral Density Beads
- d. Tufting
- e. Hydrogen Bubbles
- f. Schlieren Method

and many others. All of these methods have proven useful in a particular study of fluid flow, but in many cases are inadequate or fail to approximate the desired model. Most techniques of flow visualization require the introduction of foreign objects into the fluid which disturb the flow in such a manner as to give misleading or inaccurate results. The technique of electrochemiluminescence requires nothing except the body itself.

The technique of electrochemiluminescence, which has not been widely studied, was selected as a possible way of

investigating fluid flow behavior. Howland, Pitts, and Gesteland (2) recognized that the technique of electrochemiluminescence, if properly developed, would aid in the study of flow phenomena. With photographic considerations as a major factor, a solution was developed for maximum light intensity, the ingredients of which are listed in table 1. Springer demonstrated in (3) that the glow produced by the electrochemical reaction in laminar flow is governed by the rate of arrival of Luminol and hydrogen peroxide at the surface of the anode.

These experiments indicated that this electrochemical process could be used in two distinct ways. In the first case, the process presents a continuous display over the entire surface of the test piece where the display is in some way coupled to the flow in the boundary layer above the surface. Second, the process can be used to produce a luminous wake around the body which depicts the three-dimensional flow in the wake around the body.

The technique of electrochemiluminescence may prove useful in the study of boundary layers, transition from laminar to turbulent flow, optimizing turbulence stimulators for ship model testing, separation in two and three dimensional flow, and many other cases, limited only by

<u>SUBSTANCE</u>	<u>AMOUNT</u>	<u>PURPOSE</u>
H ₂ O	-----	Solvent
KCl	1.0 Normal	Supporting electrolyte
KOH	0.1 Normal	Adjusts pH
H ₂ O ₂	8.5×10^{-4} Normal	Oxidizing agent
Luminol*	4.4×10^{-3} Normal	Chemiluminescent substance
EDTA**	Trace	Chelating agent
Dissolved Air	Less than 50% saturation	To avoid bubbling

* Eastman Kodak Company, Luminol = (5-amino-2, 3-dihydro-1, 4-phthalazinedione)

** Eastman Kodak Company, EDTA = (Ethylenedinitrile)
Tetraacetic acid

TABLE 1. SOLUTION FOR CHEMILUMINESCENT DISPLAY

the imagination of the user.

B. Statement of the Problem

Many flow visualization techniques are seriously limited because they depend upon the inclusion of foreign devices into the stream which may disturb the flow around the test piece itself. The methods which allow visualization of flow in an undisturbed stream are few and difficult to achieve. Danckwerts and Wilson (4) experimented with a system which required the precise metering of three fluids which, when combined, react and turn color after a predictable interval of time. It is difficult to control such a reaction and the results are not always easily interpreted. The development of electrochemiluminescence as a useable technique seemed particularly attractive because of its simplicity.

Small scale experiments were performed to learn more about the electrochemiluminescent process and were successful in producing pictures of boundary layer effects, separation, and wake trails resembling the Von Karman trails using test pieces such as a flat plate, aerofoil shapes, cones and cylinders. However, the experiments were conducted using a rotating bowl which recirculated the fluid and the Reynolds number was limited to approximately 30,000. With

successful results from the pilot experiments, there remained the problem of adapting the electrochemical process to a larger scale apparatus of suitable dimensions which is capable of reaching Reynolds numbers in the range of 10^6 . With such an apparatus, it is possible to encompass the region of transition from laminar to turbulent flow as well as allowing experiments well into the turbulent regime.

C. Objectives

The objective of this project was to cover the design and construction of a suitable apparatus for use with electrochemiluminescence and capable of reaching Reynolds numbers up to 10^6 . A secondary objective was the determination of the controlling variables in the electrochemical process which were extensively studied on a small scale apparatus. The final objective of the project was to conduct a series of tests and to produce qualitative results in the form of pictures to show several fields of study to which the technique of electrochemiluminescence can be applied.

II. PROCEDURE

A. Experimental Apparatus

In order to perform a meaningful investigation in the most economic and practical fashion it was convenient to construct a small scale apparatus of the rotating bowl type pictured in figure 1 prior to construction of the full scale apparatus. The smaller machine, although limited to low Reynolds numbers, is very useful as a portable demonstration model and in this particular project proved invaluable in conducting small inexpensive tests. The larger setup, which is capable of reaching much higher Reynolds numbers, is more versatile, but, requires more time to interchange test peices than the smaller machine. A detailed description of both the small and large scale apparatus follows.

1. Small Apparatus

The small apparatus used in the experiments is pictured in figure 1. A list of equipment and their characteristics is found in Appendix A. The fluid is contained in a cylindrical plastic bowl of approximately eleven inches in diameter. Motion is imparted to the fluid by rotating the plastic cylinder by means of a system of pulleys which connects the bowl to a variable speed DC motor. The motor speed is

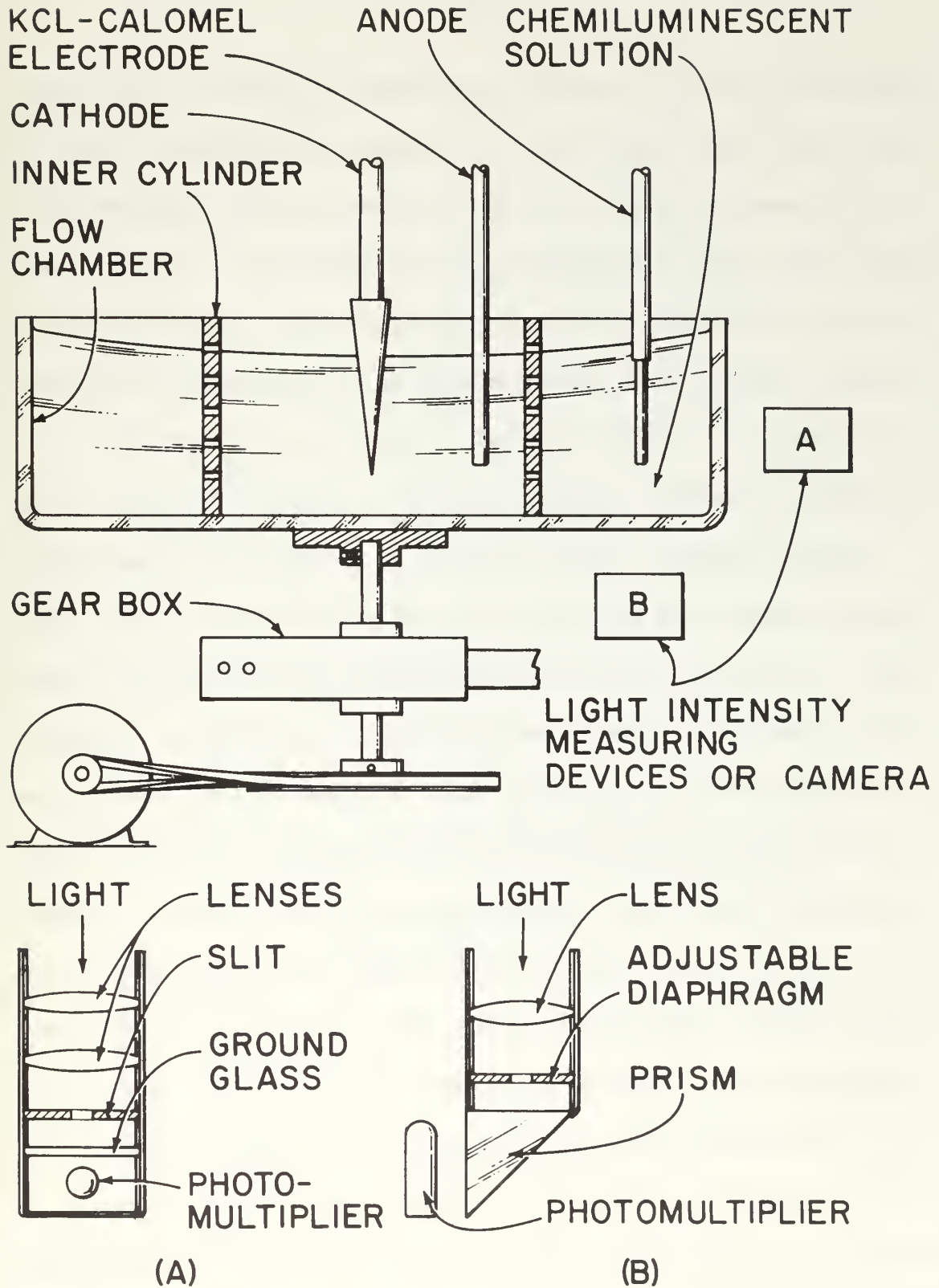


FIGURE 1. SMALL APPARATUS

controlled through a transformer rheostat which is provided to vary the Reynolds number over the range from zero to approximately 30,000. The test piece is mounted away from the center of the bowl to take advantage of the larger free stream velocity but far enough away from the outer wall to reduce the possibility of disturbances due to wall effects.

The electrical circuit, shown in figure 2, consists of a constant voltage, constant current, DC power supply with positive terminal connected to the platinum coated test piece which serves as the anode and the negative terminal connected to an aluminum section which serves as the cathode. The circuit is completed through the electrolytic solution. If the needs of the particular investigation require accurate voltage measurements between solution and anode, a vacuum tube voltmeter can be connected with positive terminal to the anode (test piece) and the negative terminal to a saturated KCl-calomel electrode located near the anode. The electrode can also be used as a reference electrode to stabilize the anode to solution potential in a "feedback" controlled circuit. However, in the type of experiments conducted in this project, no reference electrode was needed.

The rotating bowl apparatus can be used for picturing

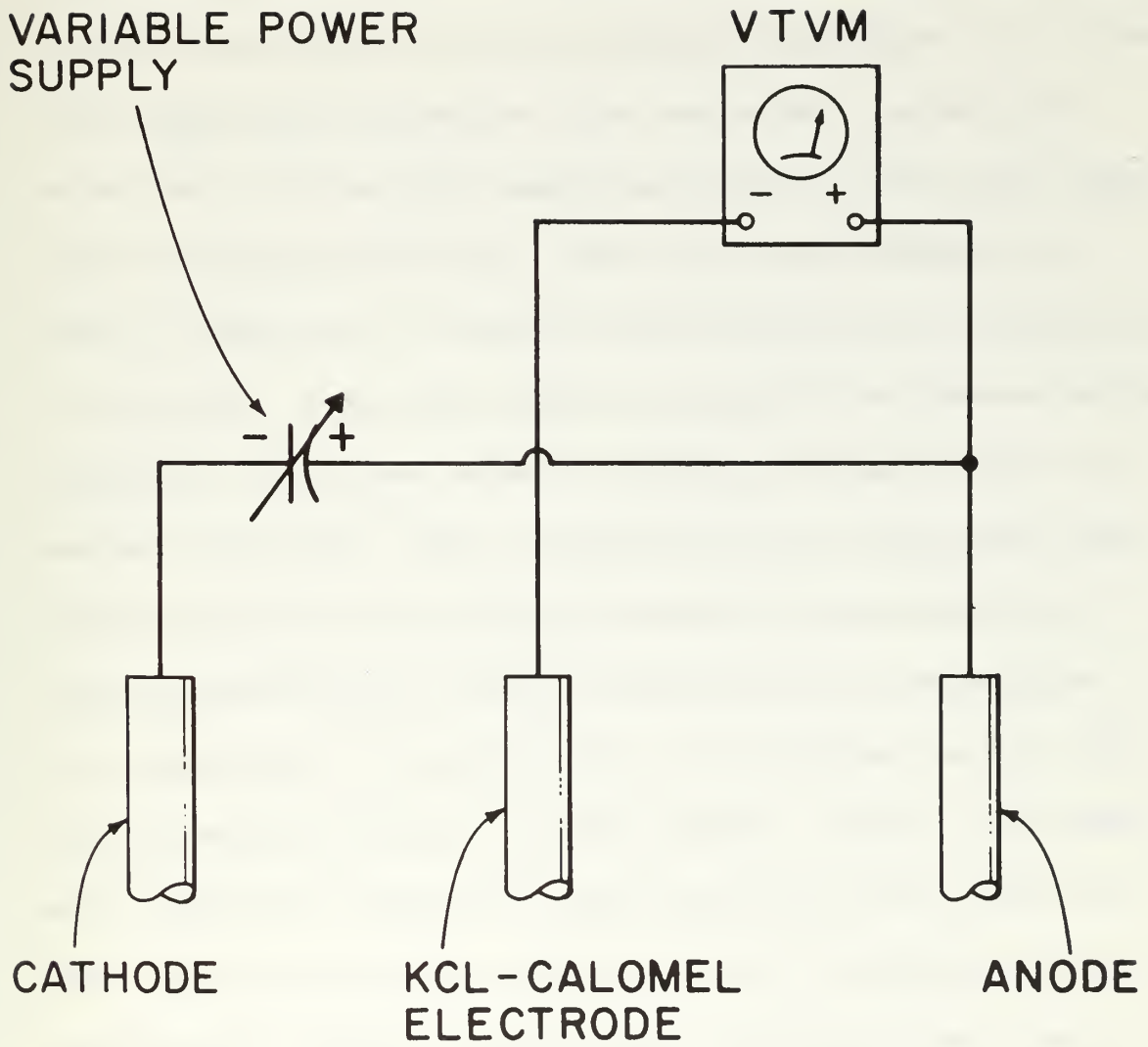


FIGURE 2 ELECTRICAL SYSTEM

a variety of low Reynolds number flow phenomena. For a detailed investigation of the governing parameters for various flow experiments, excessive time and effort was avoided using the rotating bowl apparatus because of the small quantity of solution involved. The long range usefulness of such an apparatus is found in the simplicity and portability of the small setup which makes it suitable for demonstration purposes. There are, however, several drawbacks which cannot be overlooked. The circularity of the bowl causes sufficient distortion to make an accurate optical study involving measurements, such as the position of separation on an aerofoil, of little value. In addition, we are limited in Reynolds number for two reasons. First, the higher spin velocities naturally cause the level of the fluid to rise excessively at the edge of the bowl introducing three dimensional effects in cases where two dimensional flow is desired, and second, at the higher velocities, any disturbance to the flow which is caused by the introduction of a test piece does not have sufficient time to dampen out before the particular fluid particle arrives back at the test piece again. In short, the rotating bowl apparatus is best suited for familiarization with, and the study of the technique of electrochemiluminescence as applied to flow studies

rather than the detailed flow studies themselves, except in the case of low velocity phenomena.

2. Large Apparatus

The primary objective of this project was the construction of an apparatus to use the technique of electrochemiluminescence in flow studies and reach Reynolds numbers of 10^6 maximum. A preliminary investigation was conducted in order to determine the most suitable geometry and dimensions to make a practical working system. The general layout of the apparatus and the principal dimensions are pictured in figure 3. For simplicity in discussion, the various sections will be considered separately, with tanks and associated piping first, followed by the contraction cone, the test section, and the electrical system.

a. Tanks and Associated Piping

The principal dimensions are a function of the physical problem which is being studied, the space available, and handling considerations. The physical problem considered in this report called for a Reynolds number of 10^6 so it was decided to construct a simple type of water tunnel; namely, a "blowdown" tank. The space available in the laboratory allowed the placement of the reservoir on one floor and the catch tank on the floor below with a

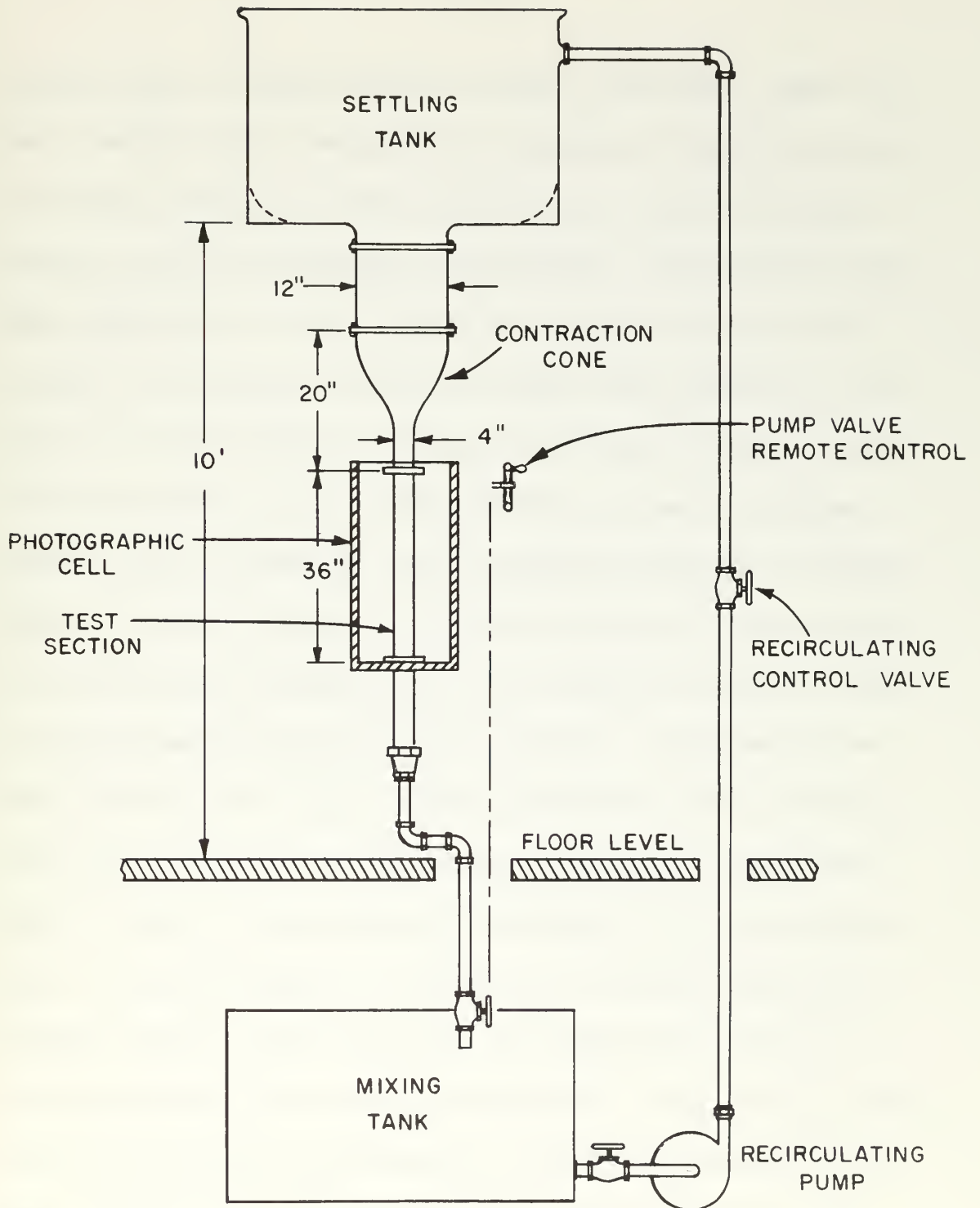


FIGURE 3 LARGE APPARATUS

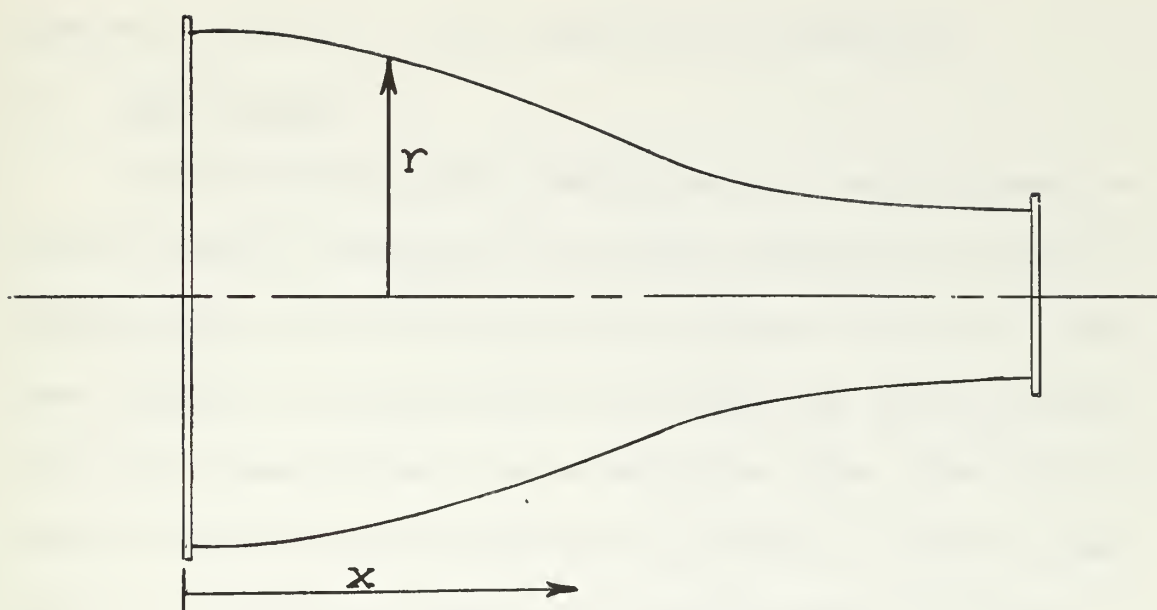
gravity head of approximately twenty (20) feet. Some of the considerations involved in arriving at the final dimensions are velocity in the test section, size of the test section, and the blowdown time desired. Calculations were made, based on an ideal fluid, for various size sections, holding the maximum velocity at that value required to obtain the length Reynolds number of 10^6 for one foot length. It was decided to use a test section of four (4) inches diameter and a blowdown time at maximum velocity of approximately one (1) minute. In order to meet the requirements of velocity and blowdown time, the reservoir capacity must be approximately fifty (50) cubic feet. The chemiluminescent fluid used in this study is alkaline and, therefore, all parts of the apparatus exposed directly to the fluid must be made of a non-corrosive material. It was decided that the tanks would be constructed of reinforced marine plywood covered with fiberglass or epoxy paint and the piping used was PVC (polyvinylchloride). The control valves and recirculating pump presented the most formidable problem. Because the main valves had to be quick-acting and non-corrosive, it was necessary to use PVC constructed valves which were very expensive. The recirculating pump is a centrifugal pump made of a thermosetting, glass

reinforced polyester, made from long strands of glass fiber and a special corrosion resistant polyester resin.

b. The Contraction Cone

The design of the channel preceding the entrance to the test section is of maximum importance because it is desired to attain a uniform velocity field free of disturbances throughout the cross section of the test section at the entrance and to have a boundary layer as small as possible. These basic requirements were achieved in an extensive design study for the design of a closed jet water tunnel (5). The contraction cone dimensions are summarized in table 2 and are a direct scaling of the original design. The rapid increase in velocity tends to decrease the boundary layer thickness and serves as a correction to velocity variations of flow provided that the preceeding and following sections have definitely established parallelism. The conditions of parallelism were achieved by the use of laboratory grade lucite tubing of twelve (12) inches diameter for the section preceeding the contraction cone and the lucite test section following the cone.

The cone was constructed by laminating pine sections and turning this mold on a lathe to achieve the desired dimensions. The pine mold was then used to build the cone of



<u>Station</u>	<u>x (inches)</u>	<u>r (inches)</u>
1.	0.00	5.999
2.	0.89	5.955
3.	1.78	5.800
4.	2.67	5.578
5.	3.55	5.133
6.	4.45	4.658
7.	5.33	4.133
8.	6.22	3.800
9.	7.11	3.444
10.	8.00	3.155
11.	8.89	2.889
12.	9.73	2.667
13.	10.67	2.497
14.	11.55	2.391
15.	12.45	2.267
16.	13.33	2.200
17.	14.22	2.133
18.	15.11	2.089
19.	16.00	2.067
20.	16.89	2.022
21.	17.78	2.000
22.	18.67	2.000
23.	19.55	2.000
24.	20.45	2.000

TABLE 2. DIMENSIONS OF CONTRACTION CONE

laminated layers of resin impregnated fiberglass.

c. Test Section

The test section was constructed of a single piece of lucite plastic tubing with an inside dimension of four (4) inches and a wall thickness of one quarter ($\frac{1}{4}$) inch. The test pieces are mounted within the tube and held in place with set screws through the side of the tube. The circular tubing is mounted within another cylinder of square cross section. The space between the two cylinders is filled with the same chemiluminescent fluid used in the test section and serves the purpose of minimizing distortion for detailed photographic analysis. Both cylinders are designed for easy removal to facilitate changing test models with minimum loss of time. If an adverse pressure gradient is desired, bleed taps may be provided in the test section and the outer cylinder will provide a drain for the excess fluid. Electrical connections are provided in the test section through the set screws. Pressure taps may be provided for velocity measurements if desired.

d. Electrical system

The electrical system is determined by the physics of the problem in that it may be as simple or as elaborate as is necessary to achieve the desired results. The system

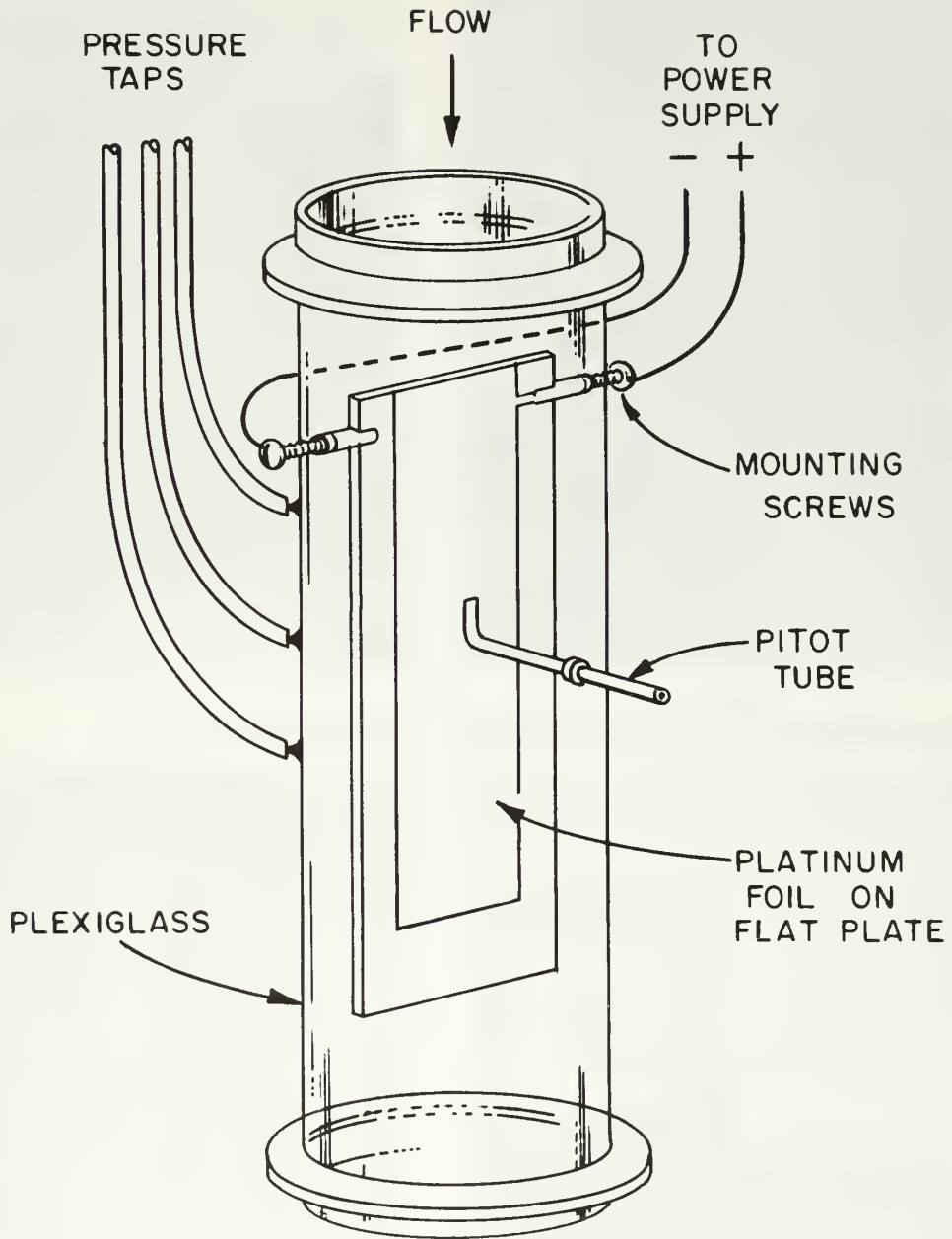
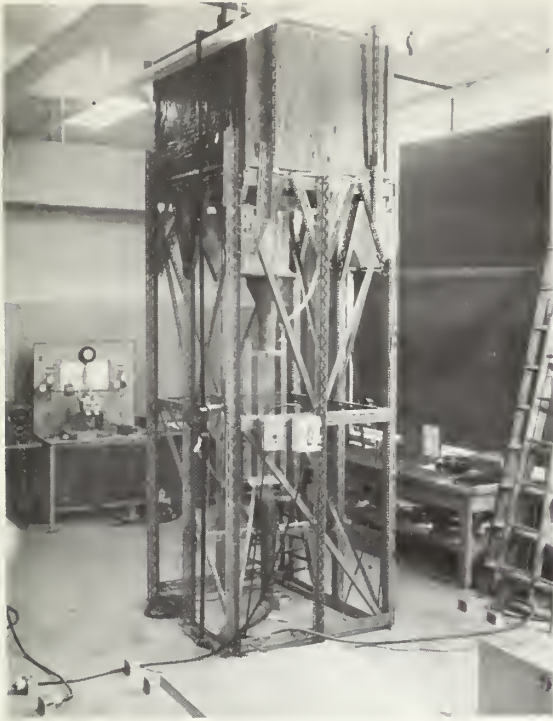


FIGURE 4

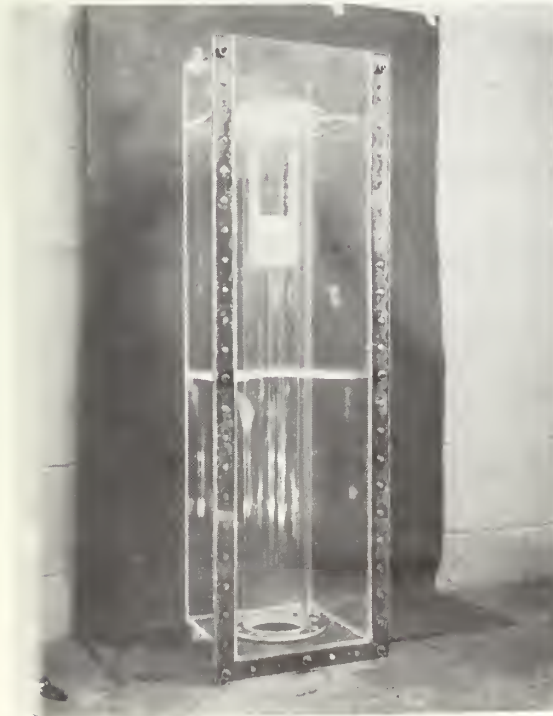
TEST SECTION



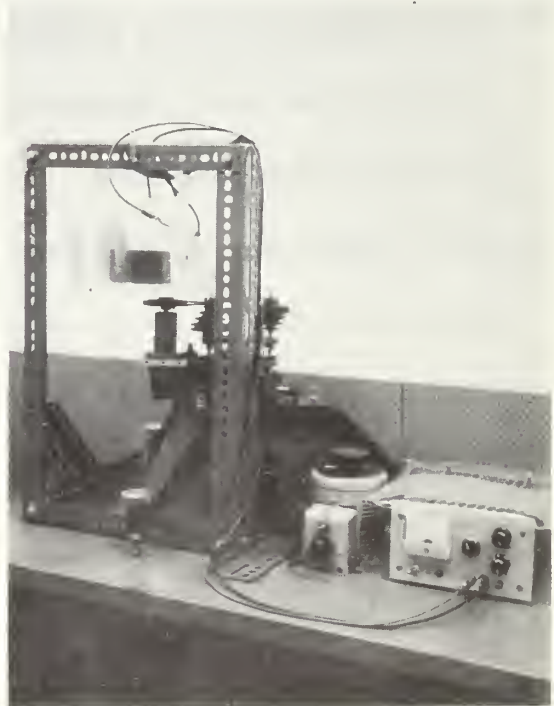
(a)
Tank, Cone and Test Section



(b)
Recirculating Pump and Piping



(c)
Test Section



(d)
Small Apparatus

FIGURE 5. COMPONENTS OF LARGE AND SMALL APPARATUS

which was used in the large apparatus for flow studies was the same as the diagram pictured in figure 2, with the exception of the KCl-Calomel electrode. The reference electrode was not necessary because the experiments were not involved in light intensity measurements or closely controlled anode to solution potentials. The power supply served as a switching device and a means of pre-setting the potential at a desired value.

The combination of all the individual components results in a system which will produce a surface glow on a body which indicates the surface effects caused by the boundary layer above the surface. The characteristics of the apparatus allow a maximum velocity in the test section of ten (10) feet per second and a corresponding blowdown time of one (1) minute. Estimated ranges of several significant quantities are shown in figures 10 and 11, Appendix C.

B. Electrochemical System

Investigations were performed in order to determine the parameters which govern the technique of electrochemiluminescence. The results of the investigation served as the guide for the selection of a proper procedure and selection of proper parameters in conducting the large scale

experiments. There are three obvious variables which were individually taken under consideration.

1. Electrodes

Initially it appeared that the anode and the cathode would have to be constructed of platinum in order to produce the proper surface glow. The use of platinum electrodes, even if the size is kept to a minimum, can be extremely expensive. The cost of the electrodes could be so high as to make it impractical to run any series of flow studies in which the flow patterns around a number of test pieces are desired. The purpose of this part of the investigation was to determine if any other materials would be suitable for use as either anode or cathode.

Various materials were tried such as aluminum and rhodium in various combinations with and without platinum. The results of the investigation indicated that platinum is the only material suitable for the anode when using a DC power supply and low values of current and voltage. It was also found that almost any material could be used as the cathode, but, satisfactory results with aluminum led to the choice of aluminum for the cathode material in all other experiments. The low cost, easy workability and availability of aluminum are also among the list of desirable

features.

There is one drawback with the use of aluminum or any material other than platinum, and that is the tendency of the cathode to cause bubbling. If the size of the cathode is maintained larger than the anode, the bubbling can be kept to a minimum. Also, the problem of bubbling is not as hindering in the blowdown type system as it is in the recirculating type system.

With the use of an inexpensive material such as aluminum for the cathode and using platinum foil to coat the test piece, the cost of the electrodes can be maintained within practical limits.

Both the electrodes must be kept clean in order to produce the maximum light attainable. The electrodes may be cleaned in a solution of potassium hydroxide in methanol when the apparatus is dismantled. If the test piece requires cleaning during a run which is evidenced by irregularities in the pattern of the surface glow or if bubbles tend to stick to the surface at low voltages, the electrodes may be electrolytically cleaned. The voltage is raised a few volts and held for a sufficient amount of time to allow the intense trails to develop and allow any bubbles to pass into the solution. With the voltage re-set to the desired

value for the particular test, the system should operate normally. Prolonged cleaning of the electrodes by electrolysis is not recommended.

2. Solution

The part of the investigation which received the most attention was the solution itself. There are several ingredients in the solution and conceivably any one or all of the ingredients could be varied and still maintain a working solution. An investigation of this type will determine what materials are necessary and in what quantity to produce a working solution. In order to have a starting point or reference point, it was decided to maintain the amount of Luminol per liter of solution constant and vary the other materials.

Since Luminol is only soluble in alkaline solutions, the amount of Potassium Hydroxide (KOH) cannot be reduced and it is of no advantage to increase the pH, as this already presents problems with corrosion. Therefore, the amount of KOH per liter of solution remained unchanged.

The next substance to consider is the Potassium Chloride (KCl). This ingredient is added in large amounts and serves as the supporting electrolyte. With large quantities of solution such as are used in the blowdown tank,

the amount of potassium chloride is cumbersome and expensive. Tests were run to see if the amount of KCl could be substantially reduced while still maintaining a working solution. As a result of the tests, it was found that the concentration of KCl could be reduced to one-third of the concentration listed in table 1. without any noticeable effect on the properties of the solution. This cut the cost of an expensive item in the solution by two-thirds of the original cost.

The hydrogen peroxide, which is the oxidizing agent, can be varied depending on the desired life of the solution. We were trying to get the most useful time out of a particular batch of solution so it was decided to maintain the hydrogen peroxide at the recommended level. If it becomes necessary to reactivate the solution, additional hydrogen peroxide can then be added.

It is recommended that the dissolved air be reduced to at least 50% of the saturation level. For small quantities of solution, it is easy to vacuum boil the solution in a vacuum container. However, when dealing with large quantities of solution and large wall sided containers it is almost impossible or at least very impractical to vacuum boil the solution. Other methods of degassing would present

similar problems so it was decided to attempt to run without degassing the solution. Satisfactory results were obtained without degassing which is attributed to the fact that a blowdown apparatus rather than a recirculating device was used.

The last two ingredients, namely EDTA and water, are discussed together as the use of the former depends on what type water is used. Impurities in the water, such as copper or iron, will produce an objectional bulk glow in the solution, so, if such impurities exist in the water, a small amount of EDTA is added to precipitate out the metallic impurities. If distilled water is used, no EDTA is necessary. Distilled water presents a problem in procurement in large quantities for the blowdown apparatus. The addition of EDTA is time consuming because tests must be run each time a solution is mixed to determine the amount of EDTA to add. As an alternate means of producing water which is free of metallic impurities, tap water was passed through ion exchange cartridges which removed all the impurities and produced water with conductivity comparable to that of distilled water. This system appears to provide water with the desired properties in the cheapest and most convenient manner.

The basic solvent may be changed to produce some interesting results. For example, glycerin is added to adjust the kinematic viscosity and to slow down the reaction for longer photographic exposures. The glow produced does not seem to be affected by the addition of solvent materials such as methanol, acetone, and others described in (2).

The lifetime of such a large quantity of solution is of importance because the amount of chemicals used in preparing a batch of solution makes it costly. A list of cost estimates is found in Appendix B.

The electrochemical solution changes with time which is evidenced by decreased light output and discoloration. Experiments related to the investigation of heat or mass transfer are strongly dependent on light intensity and therefore, the solution is good only as long as the voltage to light output relationship remains constant. Howland, Pitts, and Gesteland indicate in their report that the voltage to light output relationship remains constant for approximately twelve (12) hours.

On the other hand, if the type of investigation is simply a visual study, such as determining the relationship between the position of separation and the angle of attack

and Reynolds number, the useful lifetime of the solution is considerably longer than when it is used for light intensity measurements. Solutions were used successfully for periods up to seven (7) days without adding anything to revitalize the solution. During the seven day period, sufficient light was developed to allow photographing of the flow patterns.

The hydrogen peroxide is the first chemical which is used up because it is constantly breaking down during the time that a potential is applied between anode and cathode, and since the components do not recombine, the hydrogen peroxide is eventually depleted. If desired, small amounts of hydrogen peroxide can be added to the solution from time to time in order to reactivate the solution.

3. Voltage

Throughout the experiments, a check was made in order to determine the relationship between voltage and Reynolds number, if any. For the complete range of Reynolds numbers, the peak light intensity occurred at approximately the same voltage which was 2.5 to 3.0 volts measured from anode to cathode. This independence from Reynolds number is substantiated by Springer (3) and the results of his experiments are shown in figure 6. The current limiting process

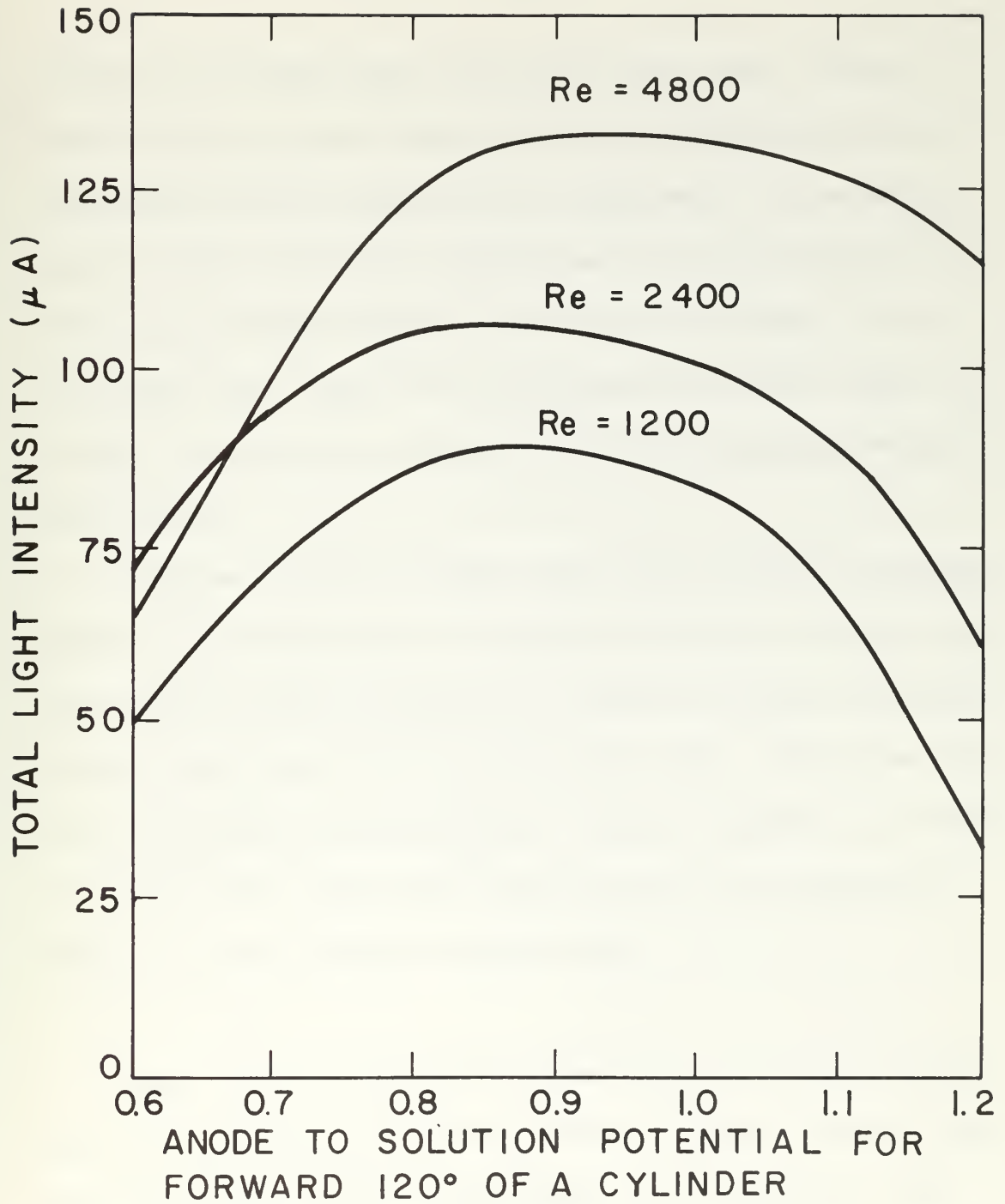


FIGURE 6 PLOT OF LIGHT INTENSITY VS POTENTIAL

causes the maximum light output to occur over a fairly broad voltage range, which eases the problem of voltage adjustments while conducting an experiment. Fine adjustments in this band will control the contrast while the total light output remains essentially unchanged. The increased light output with increasing Reynolds number is readily evident even without the aid of intensity measuring devices.

The glow pattern changes when the voltage is increased beyond that value which corresponds to maximum light output. At slightly higher voltages, the light intensity decreases rapidly until the glow is practically gone. When the voltage is increased still further, the glow appears in streams or trails which break away from the body and apparently follow the wake produced by the body.

This investigation has covered the controlling parameters for performing a series of flow studies and has indicated that the items of importance are as follows. The anode must be platinum or platinum plated and in the shape of the body around which the flow pattern is desired. The cathode may be of any material as long as the size is sufficient to avoid excessive bubbling. Corrosion problems

should also be considered in the selection of the cathode. The composition of the solution can be varied greatly without substantially affecting the light output or the flow pattern. For maximum light output, the solution listed in table 1 is recommended.

There are several questions that are still to be answered. Although it is evident that the proper selection and setting of the controlling variables on a particular test piece will produce a glow, it is not readily evident just what chemistry causes the glow. Finally, it must be determined what this glow can be used for in the study of fluid mechanics.

III. FLOW VISUALIZATION

It has been demonstrated that the proper selection of parameters, as outlined in the previous sections, will produce a glow on the surface of a body. The glow intensity is governed by the rate of arrival of reactants as demonstrated in (2). An attempt has been made to uncover the mechanism that causes the glow. With a general understanding of what is happening, the flow patterns can be interpreted. Specific studies such as separation of flow and tripping from laminar to turbulent flow were used as examples of possible applications of the technique of electrochemiluminescence.

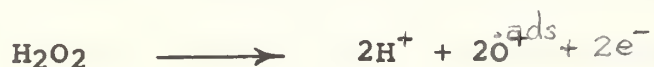
A. Electrochemical Process

The electrochemical process, i.e., the use of an electric current to trigger a chemical reaction of substances in solution to produce a glow, is not original. However, all the questions related to the chemistry of the particular reaction resulting with a solution containing Luminol have not been answered. The experience, which was gained from repeated experiments, provided a partial explanation of the mechanism by which the chemiluminescent glow is produced. The explanation will be straightforward and brief in the form of a hypothesis. Further experiments will be

needed in the future to continue the study of the electrochemiluminescent process.

In his paper on the use of electrochemiluminescence, (3) Springer has stated and verified by experiments that the bluish glow is governed solely by the rate of arrival of reactants at the anode in laminar flow. The process was also catagorized as a "limiting current" condition, where the light intensity increases with an increase in voltage and reaches a maximum. As shown in figure 6, further increase in voltage causes the light intensity to decrease. Still higher voltages cause streamers or trails to leave the body and follow the wake.

Let us say that the blue glow in the solution is produced when dissolved Luminol molecules unite with free oxygen ions. The Luminol molecules are in abundance in the solution so the glow is dependent only on the production of oxygen ions with which the Luminol molecules can then react. The platinum anode with a low potential applied provides the stimulus for producing the oxygen ions. Hydrogen peroxide (H_2O_2), at the anode, breaks down according to the following reaction;



and the oxygen ions then combine with Luminol molecules, also present at the anode, and produce a surface glow.

The three mechanisms by which the substances can arrive at the anode are:

- a. Migration of ions
- b. Diffusion
- c. Convection

We are interested in the uses of this process in the study of fluid flow phenomena and therefore, want to avoid any reaction caused by the migration of ions due to potential difference. The addition of potassium chloride (KCl) increases the conductivity of the solution and thereby reduces the possibility of ion migration. Diffusion and convection remain as the means of transporting material to the anode and both these mechanisms govern the arrival rate of reactants at the anode as brought out in reference (3).

In order to acquire a flow picture which is representative only of the disturbances caused by the introduction of the body into the fluid, or for mass transfer studies, the principle of convection, either natural or forced, should be the only means of transporting material to the surface of the anode. However, in the electrochemical solution, convection and diffusion are inseparable. If

it can be determined, apriori, that one or the other predominates, experiments will provide meaningful results. In the study of separation, the free stream velocity was sufficiently large that diffusion effects were negligible. In cases where diffusion effects become more important, such as in extremely low velocity studies, the results of the visual study must be considered more carefully to separate the two effects before drawing any conclusions.

At low voltages, a small amount of hydrogen peroxide is broken down at the anode, thus forming oxygen ions which react immediately with the Luminol to produce a faint glow. As the voltage is increased, more hydrogen peroxide is broken down, and a greater number of ions are available to react with the Luminol producing a brighter glow. When the maximum light intensity is reached, the voltage is sufficient to cause all the hydrogen peroxide arriving at the anode to break down, thereby reducing the concentration of hydrogen peroxide at the surface to zero. Therefore, a further increase in voltage cannot produce any additional oxygen ions from hydrogen peroxide in steady flow.

The surface reaction is the mayor topic under discussion but something interesting occurs at higher voltages which can possibly be more completely explained by continued

experimentation. When the voltage is increased beyond the value which produces maximum light intensity, there is a sudden reduction of the light intensity at the surface and if the voltage is increased still further the blue glow is observed to leave the surface in streamers or trailers and produce a three dimensional effect in the wake. It is conceivable that at a voltage slightly above that for maximum light intensity, oxygen ions are combining to form molecular oxygen which causes polarization of the platinum anode and thus prevents hydrogen peroxide from reaching the surface. At still higher voltages, the water itself may break down in the vicinity of the anode and produces the ions which, when carried away from the surface, react with Luminol in the free stream or wake.

B. Separation on a Flat Plate

The object of the first series of tests was to place a flat plate in a uniform stream at a fixed angle of attack equal to three degrees and to vary the free stream velocity from zero to approximately six feet per second with a corresponding range of Reynolds numbers from zero to approximately 200,000. This series of tests was to show the variation of the position of separation with varying Reynolds numbers at a fixed angle of attack. The

pictures in figure 7 clearly show that the position of separation is easily identified by the abrupt change in light intensity which is caused by the radical difference in velocities in the separated and unseparated areas. Figure 7(a) shows that at low Reynolds numbers, the flow is unseparated. Figures 7(b), (c), and (d) show that as the Reynolds number is increased sufficiently, separation occurs and for a further increase in Reynolds number, the position of separation moves toward the leading edge of the plate as explained in (7).

The pictures obtained from the second series of tests are shown in figure 8. The free stream velocity was the same in all these runs and the angle of attack of the flat plate was varied from zero to seven degrees. At zero angle of attack, the flow is completely laminar and produces a uniform glow shown in figure 8(a). As the angle of attack is increased, separation occurs and figures 8(b), (c), and (d) show that for increasing angles of attack, the position of separation moves towards the trailing edge of the plate which is in accordance with existing theory. The curved streaks in figure 8(d) were caused by the flow of fluid from the high pressure side of the plate to the low pressure side around the edges of the plate



(a)
Rey = 10,000



(b)
Rey = 60,000



(c)
Rey = 100,000



(d)
Rey = 200,000

FIGURE 7. SEPARATION ON A FLAT PLATE AT VARIOUS REYNOLDS NUMBERS (BASED ON PLATE LENGTH OF 4.75"). ANGLE OF ATTACK=3°



(a)
Angle of attack = 0°



(b)
Angle of attack = 3°



(c)
Angle of attack = 5°



(d)
Angle of attack = 7°

FIGURE 8. SEPARATION ON A FLAT PLATE AT VARIOUS ANGLES OF ATTACK. REYNOLDS NUMBER (FOR PLATE LENGTH OF 4.75")=100,000

C. Turbulence Stimulators

Models of full scale ships are towed and the resistance is measured and scaled to determine the resistance of the full scale ship. The full scale resistance determines the horsepower required for the ship, so estimates should be as accurate as possible. Models are towed at speeds which produce laminar flow around the hull of the model. In order to arrive at the best possible resistance estimates, the flow around the model must be turbulent. Several methods have been used to trip the laminar flow into turbulence which include trip wires, plate studs, cylindrical studs, triangular pyramid studs, nail studs, and others (8). Very little data exists regarding the critical size and the proper placement of the various types of turbulence stimulators. The present methods of investigation of the effectiveness of the various stimulators include the hot wire anemometer, the benzoic-acid film method and white milk injection. The small amount of data produced by the existing techniques suggests the need for new and better methods.

An attempt was made to show the effect of the placement of stimulators by gluing cylindrical studs of the type currently used by the Ship Model Towing Tank at

Massachusetts Institute of Technology on a flat plate at different distances from the leading edge of the plate. The free stream velocity was set at three feet per second to approximate the towing speed of models less than five feet in length. The studs, which are one-eighth of an inch in diameter and one-tenth of an inch high, produced the flow pattern in figure 9. The studs nearer the leading edge are not effective but those further back on the plate succeed in producing turbulent flow.



FIGURE 9. FLOW PAST A FLAT PLATE FITTED WITH
TURBULENCE STIMULATORS. FREE STREAM
VELOCITY = 3.0 FEET PER SECOND.

IV. CONCLUSIONS

The tests conducted with the apparatus under consideration have shown that the technique of electrochemiluminescence can be used in the study of boundary layers. The pictorial results shown in figures 7, 8, and 9 verify the usefulness of the technique of electrochemiluminescence in studies of separation of flow and the development of turbulence stimulators.

The prime advantage of this technique is the fact that a boundary layer display of every point on a body can be obtained without the need of objectional devices which produce undesirable flow disturbances.

It has also been shown that this type of apparatus can be used to apply the technique of electrochemiluminescence to heat and mass transfer studies.

With such a broad range of applications, the technique of electrochemiluminescence can be added to the family of Flow Visualization Techniques in the study of Fluid Mechanics.

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APPENDIX A
LIST OF EQUIPMENT

1. Power supply, Harrison Laboratories, Model number 855B;

Input: 105-125 volts AC, single phase, 50-440
cycles

Output: 0-18 volts, 0-1.5 amps., constant voltage
or constant current

Size: 7 $\frac{3}{16}$ " wide, 5 $\frac{1}{16}$ " high, 8 $\frac{1}{2}$ " deep

2. Rotating bowl drive motor, Robbins and Meyers,

"The Standard Motor"

Input: 110 volts AC, single phase, 60 cycles

Output: 0-6000 R.P.M.

3. Motor speed controller, The Superior Electric Company
Inc., "Powerstat", Type 116

Input: 120 volts AC, single phase, 50-60 cycles

Output: 0-140 volts

4. Pump drive motor, General Electric, TriClad induction
motor, Model number 5K182AG1

Input: 209-220/440 volts AC, 3 phase, 60 cycles

Output: 1 $\frac{1}{2}$ Horsepower @ 3495 R.P.M.

5. Recirculating pump, Deming Division, Crane Co., "Fianite"
centrifugal pump; 1 $\frac{1}{4}$ " inlet, 1" outlet, 55 G.P.M.
capacity for 70 foot head @ 3500 R.P.M.

APPENDIX B
COST OF CHEMICALS

1. Potassium Chloride Reagent

KCl (bulk) \$0.445/pound

2. Potassium Hydroxide Reagent

KOH \$1.22/pound

3. Hydrogen Peroxide Reagent

H₂O₂ 30% Solution \$2.67/pint

4. Luminol \$10.30/25grams

5. Ion Exchange Cartridges \$12.00 each

APPENDIX C
SYSTEM PARAMETERS

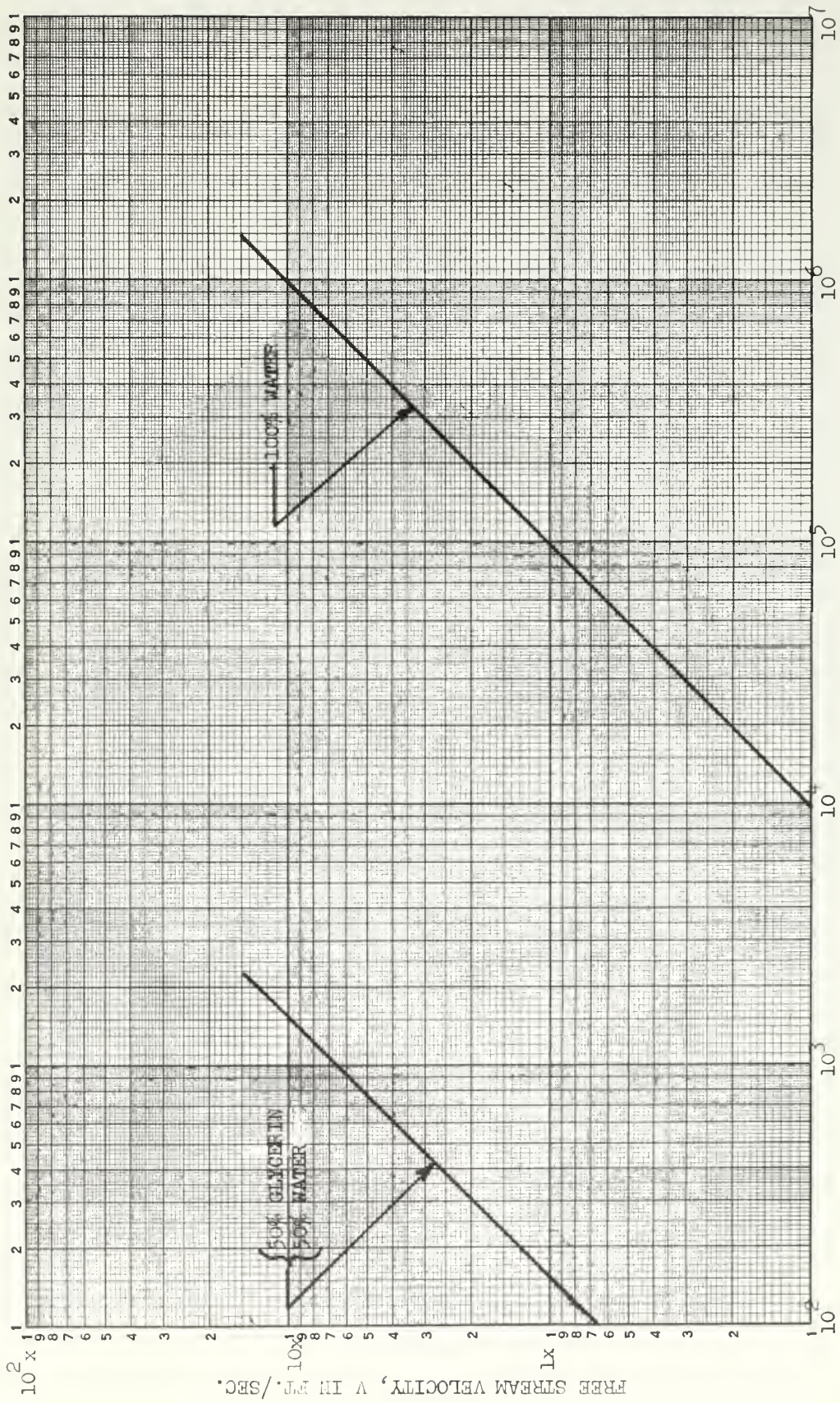
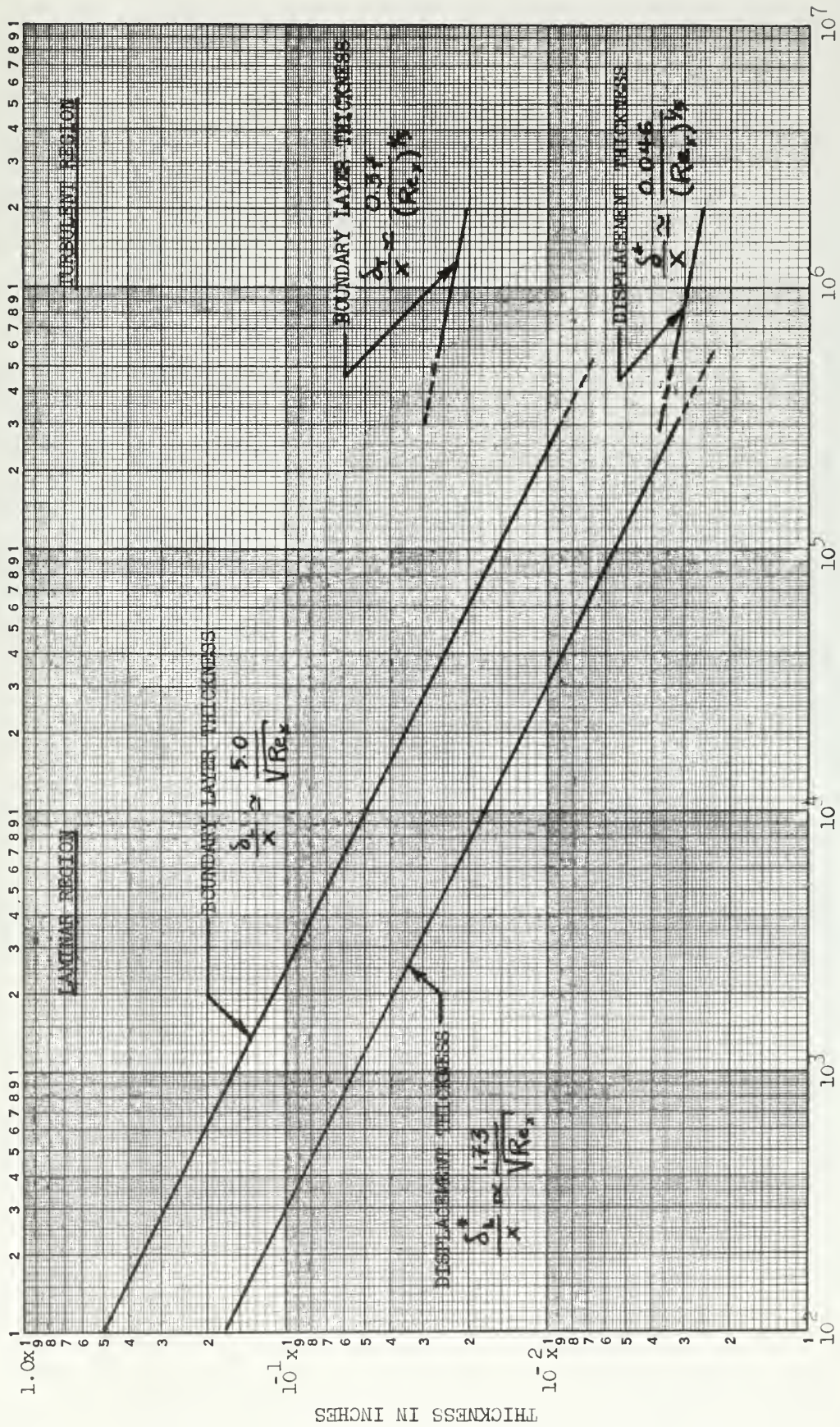


FIGURE 10. ESTIMATED RANGE OF FREE STREAM VELOCITY IN TEST SECTION

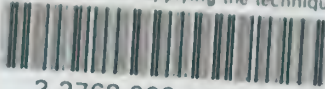


REYNOLDS NUMBER (BASED ON $L = 1.0$ FEET)

FIGURE 11. ESTIMATED RANGE OF BOUNDARY LAYER AND DISPLACEMENT THICKNESS IN TEST SECTION

the 5367

An apparatus for applying the technique



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